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Magnetic field-dependent photoluminescence linewidths as a probe of disorder length scales in quantum wells

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Photoluminescence from highly disordered GaAs quantum wells is studied in magnetic fields up to 50 T. The monotonic decrease of the photoluminescence linewidth with increasing quantum well thickness indicates that interface roughness is the primary source of line broadening. The magnetic field-dependent exciton linewidth shows an unexpected behavior. We observe not only just a monotonic increase in linewidth but also a field-dependent decreasing linewidth in thicker quantum wells. These observations are understood by postulating the existence of two correlation lengths for the interface fluctuations, one much smaller than the exciton size and the other one of the order of the exciton size. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2825417]

Most of the recent advances in semiconductor physics and technology have been a direct consequence of our ability to confine electrons between a pair of heteroepitaxial interfaces. Since the physical interfaces can never be absolutely smooth, understanding the relationship between the electronic wave functions and the imperfections of the interface continues to be an active topic for research.[1–6]

Excitonic emission as seen in photoluminescence (PL) spectra can be inhomogeneously broadened by quantum well (QW) thickness fluctuations. If the QW thickness L_z dependent transition energy $E_T(L_z)$ obeys an effective mass-type relationship, $E_T(L_z)=E_g+AL_z^{-\alpha}$, where A and α are two fitting parameters and E_g the bandgap, then the emission linewidth σ is expected[2] to scale as $\sigma \propto L_z^{-(\alpha+1)}\Delta L_z$. This dimensional argument suggests that if ΔL_z, the magnitude of interface fluctuations, is independent of the well width L_z, the effective *strength* of the interface disorder is inversely dependent on the well width. One may refine this argument somewhat to include lateral correlations in the disorder potential,[2,3] provided that the correlation length χ_1 of the interface disorder potential is much smaller than the averaging volume (exciton size a_B). In the context of high quality heterostructures, χ_1 is associated with interface fluctuations on the monolayer scale. These monolayer fluctuations will exist even without any growth nonuniformity and are an inevitable consequence of having a semiconductor alloy forming the barrier region. For example, in $Al_{0.3}Ga_{0.7}As/GaAs/Al_{0.3}Ga_{0.7}As$ QWs, the barrier itself is a random $Al_{0.3}Ga_{0.7}As$ alloy and Ga–Al bonds only occur in 30% of the cases. Thus, the notion of a sharp interface, inspired by a virtual crystal picture, is naive. Monolayer and larger, random fluctuations must always exist. The excitonic linewidth due to these fluctuations[2] is given by

$$\sigma \propto \frac{\partial E_T}{\partial L_z}\left[\frac{\chi_1}{a_B}\right] \propto \frac{\Delta L_z}{L_z^{\alpha+1}}\left[\frac{\chi_1}{a_B}\right], \quad \chi_1 \ll a_B. \qquad (1)$$

The $[\chi_1/a_B]$ factor, which considerably reduces the effect of these fluctuations on the exciton, is simply the consequence of the $1/\sqrt{N}$ law for the second moments of a random variable.[3] Physically, this disorder averaging is another manifestation of the well-studied motional narrowing phenomenon.[3] In the past, this sensitivity to the exciton size has been quite spectacularly demonstrated in magnetic field B dependent PL measurements. Many groups have observed a field-dependent enhancement in the emission linewidth. It follows from Eq. (1) that the squeezing of the exciton wavefunction would lead to a reduced disorder averaging[7–9] and, consequently, an increase in the PL linewidth.

Interestingly, this is completely contradictory to some earlier measurements[10,11] where the linewidth was observed to decrease as a function of magnetic field. These old observations have largely been ignored and the considerable theoretical and experimental work on the subject in the last two decades has only focused on the magnetic field-induced enhancement of the emission linewidth.

We have revisited this problem in the present study. Specifically, we will discuss a GaAs multi-QW sample where the disorder-induced inhomogeneous broadening is about an order of magnitude larger than the homogeneous broadening for GaAs QWs. Our paper describes results of PL measurements on such QWs in high magnetic fields up to 50 T.

Four independent $Al_{0.3}Ga_{0.7}As/GaAs/Al_{0.3}Ga_{0.7}As$ QWs with thicknesses of 20, 30, 50, and 100 Å were contained within a single sample. The QWs were grown in a horizontal

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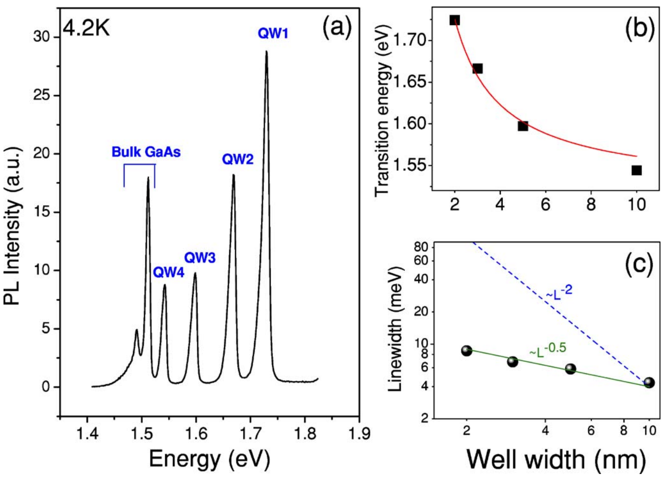

FIG. 1. (Color online) (a) Photoluminescence spectrum of the GaAs multi-QW sample measured at 4.2 K and $B=0$. Four peaks corresponding to the four quantum wells are clearly seen. (b) Variation of the peak emission energy with the QW thickness. The solid line corresponds to a curve with $\sim L^{-1}$. (c) Linewidth as a function of well width. The solid line corresponds to a curve $\sim L^{-0.5}$. One would naively expect an $\sim L^{-2}$ dependence (broken line) since the PL energy has L^{-1} dependence.

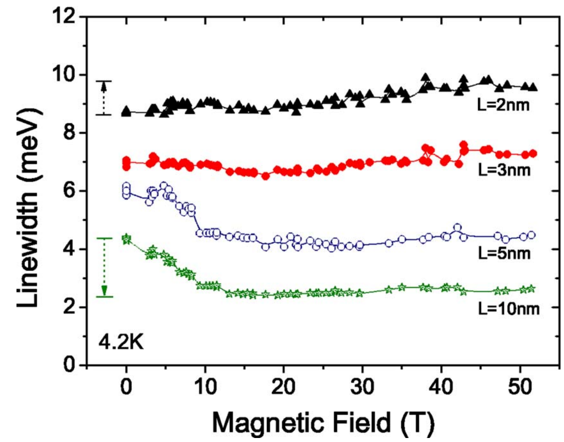

FIG. 2. (Color online) Change in the linewidth, $\sqrt{\langle E^2\rangle - \langle E\rangle^2}$, as a function of magnetic field for quantum wells of different thicknesses. Qualitative trends depend on the well width.

metal-organic vapor phase epitaxy reactor at a nominal rate of 4.8 Å/s using trimethylaluminum and trimethylgallium as the group-III sources and arsine as the group-V source. The QWs are uncoupled due to a 1300 Å thick $Al_{0.30}Ga_{0.70}As$ barrier separating each well. Since all the QWs are in the same sample, the other sample characteristics, in particular, the background impurity levels, can be assumed to be constant across the QWs.

Nonresonant PL measurements were performed in a liquid helium bath cryostat using a green diode-pumped frequency-doubled neodymium-doped yttrium aluminum garnet laser in pulsed fields up to 50 T. The spectra were measured using an intensified charge-coupled device detector. Figure 1 describes the PL measurements done at 4.2 K and $B=0$. In Fig. 1(a), individual peaks from the four QWs and the GaAs substrate are separately resolved. The spectral lineshapes are asymmetric, with the asymmetry of the peaks inversely related to the well thickness. This is most probably due to contributions from free and interface fluctuation-bound excitons.[7] However, in the absence of a clear splitting of the QW emission spectrum into two distinct peaks, the whole spectrum is better treated as a composite entity. Therefore, instead of making assumptions about the spectral lineshape and, for example, fitting multiple Gaussians or more complex Lorentz-Gaussian lineshapes[8] to a PL spectrum $I(E)$, a direct and fitting-parameter-free interpretation of the spectra is obtained by relating the peak emission energy and the linewidth to the first moment ("center of mass" $=\langle E\rangle$) and the second moment of energy ($\sigma=\sqrt{\langle E^2\rangle - \langle E\rangle^2}$). Here, $\langle E^n\rangle = \int E^n I(E)dE / \int I(E)dE$. The use of the term linewidth in this paper refers to this standard deviation σ, which for a Gaussian lineshape is, of course, $[1/(2\sqrt{2\ln 2})]\times$ the full width at half maximum (FWHM).

Figures 1(b) and 1(c) show the PL energies and the linewidths corresponding to the four QWs as a function of well width. An inverse relationship between the PL linewidth and QW thickness [Fig. 1(c)] suggests an important role of interface roughness in determining the exciton broadening. However, the actual dependence of the linewidth on the well width [Fig. 1(c)] is found to be $\sigma \sim L^{-0.5}$ and is much weaker than the expected $\sigma \sim \partial E_T/\partial L_z \sim L^{-2}$ [since $E_T \sim L^{-1}$ from Fig. 1(b)]. This suggests that $[\chi/a_B]$ is not constant for the different QWs.

Figure 2 shows the variation of the PL linewidths of the four QWs as a function of magnetic field. The expected[2,9] increase in the linewidth with magnetic field is observed only in the thinnest QWs, where the linewidth increases by about 10% in a field of 50 T. From Eq. (1), this corresponds[9] to a shrinkage of the excitonic radius by the same amount. The 10 and 5 nm thick QWs, on the other hand, show a clear *decrease* in the linewidth. In fact, there seems to be a gradual crossover from the field-induced decrease to a field-induced increase in the emission linewidth as we go from the thick to the thin QWs. As mentioned earlier, the observations of the decrease in linewidth, from the point of view of more recent theories, seem to be quite unexpected. However, they do have a very striking resemblance to the two decade old data of Sasaki *et al.*[10] on QWs with similar linewidths. Unfortunately, Sasaki *et al.* did not provide a satisfactory explanation for the observations. The field-induced decrease in linewidth was attributed to the suppression of homogeneous broadening (phonon) processes, whereas it is now quite well established that the FWHM due to phonon scattering is only[8] of the order of 0.5 meV.

Since there is no theoretical framework to understand the magnetic field-induced suppression of PL linewidth, we shall outline a plausible physical mechanism. It is likely that, along with the inevitable short-ranged monolayer alloy fluctuations,[3] the QW may also have longer ranged "real" well-width fluctuations. This would also explain the large linewidths in our samples. [For example, the molecular-beam-epitaxially-grown 2 nm QW sample measured in Ref. 5 has a linewidth of 3 meV (FWHM=7 meV) compared to 8 meV in our case.] The roughness of the interface would be very dependent on the conditions prevailing in the reactor at the time of growth. For instance, the actual morphology of the growing surface is known to be very sensitive to the growth kinetics parameters; higher growth rates and lower growth temperatures lead to growth of rough surfaces. We postulate that such fluctuations occurring on a length scale $\chi_2 \sim a_B$ are the primary source of disorder in our QWs. These longer ranged fluctuations would be expected to carry the correlations across the other interface over much larger well

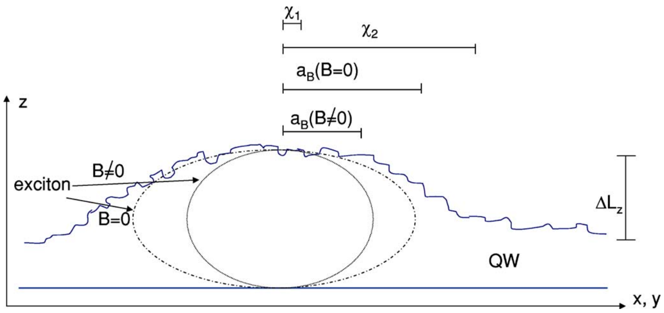

FIG. 3. (Color online) A schematic of the probable physical situation. The figure depicts quantum well interface fluctuations on two length scales, $\chi_1 \ll \chi_2 \sim a_B$. The other interface is drawn flat for simplicity.

widths and thus, unlike the local height fluctuations which were expected to be constant across the QWs, global fluctuations would preferentially affect the thicker wells more than the thinner wells. This is also supported by the observation in Fig. 1(c), where the well-width dependence of the linewidth had a weaker dependence of the inverse well-width than would be expected from the dimensional arguments of Eq. (1).

To see how the introduction of a larger correlation length can lead to the observed field-induced decrease in linewidth, consider the following simple argument: The global fluctuation of the QW thickness can be assigned an in-plane position-dependent effective potential, $V(x,y) = \partial E/\partial L_z \langle \delta L_z(x,y)\rangle_{a_B}$, where $\delta L_z(x,y)$ is the variation over the mean thickness at the in-plane position coordinate (x,y). The angular brackets $\langle \cdots \rangle_{a_B}$ denote averaging over the exciton size. We assume that this fluctuation has the form $\delta L_z(\rho) = A\exp[-\rho/\chi_2]$, where $\chi_2 \sim a_B$ is the length (measured in the plane of the quantum well) over which the fluctuation is active and ρ is just the transformation from cartesian to in-plane circular coordinates. Then, the broadening is just proportional to the square of this fluctuation averaged over the exciton size; i.e., the linewidth as a function of the exciton size can be written as

$$\sigma_2 \propto \frac{\partial E}{\partial L_z}\int_0^{a_B} \exp[-\rho/\chi_2]d\rho = \frac{\partial E}{\partial L_z}[1 - e^{-a_B/\chi_2}]. \quad (2)$$

The above expression predicts a decrease in the linewidth as the excitonic Bohr radius is shrunk in the applied magnetic field ($a_B \sim 1/\sqrt{B}$). In the limit of the correlation length being much larger than the exciton, Eq. (2) reduces to

$$\sigma_2 \propto \left[\frac{\partial E}{\partial L_z}\right]\left[\frac{a_B}{\chi_2}\right] \quad \text{for } \chi_2 \gg a_B, \quad (3)$$

which is in complete contrast with Eq. (1).

The above back-of-the-envelope derivation is supported by the qualitative picture shown in Fig. 3. In a magnetic field, the excitonic wave function would reduce in size and, therefore, encounter less of the fluctuating interface. For fluctuations which vary on a length scale comparable to the excitonic size, this would lead to a decrease in the linewidth. A reduced averaging of the local monolayer fluctuations would, on the other hand, increase the exciton linewidth. This gives rise to the competing behavior observed in the four samples (Fig. 2). It is interesting that the thinner QW with a larger linewidth seems to be more sensitive to the small scale potential fluctuations. This is tentatively attributed to much stronger exciton localization in the thinner wells, due to the larger effective disorder $(\delta L/L)$ and linewidth, and because the thinner QW is closer to being an ideal two-dimensional system.

In summary, we have studied the magnetophotoluminescence spectra of highly disordered quantum wells with intermediate thicknesses of 20 Å $< L_z <$ 100 Å in magnetic fields up to 50 T. The linewidth associated with excitonic emission shows a complex behavior, it can either decrease or increase in the magnetic field. The field-induced decrease in linewidth was attributed to an inhomogeneous broadening contribution from an interface disorder potential that varies on the scale of the exciton Bohr radius.

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[1] M. A. Herman, D. Bimberg, and J. Christen, J. Appl. Phys. **70**, R1 (1991).
[2] K. K. Bajaj, Mater. Sci. Eng., R. **34**, 59 (2001); J. Singh, K. K. Bajaj, and S. Chaudhuri, Appl. Phys. Lett. **44**, 805 (1984); J. Singh and K. K. Bajaj, J. Appl. Phys. **57**, 5433 (1985).
[3] E. Runge, *Solid State Physics*, edited by H. Ehrenreich and F. Spaepen (Academic, San Diego, 2002), Vol. 57, p. 150.
[4] F. Natali, D. Byrne, M. Leroux, B. Damilano, F. Semond, A. Le Louarn, S. Vezian, N. Grandjean, and J. Massies, Phys. Rev. B **71**, 075311 (2005).
[5] M. Erdmann, C. Ropers, M. Wenderoth, R. G. Ulbrich, S. Malzer, and G. H. Döhler, Phys. Rev. B **74**, 125412 (2006); C. Ropers, M. Wenderoth, L. Winking, T. C. G. Reusch, M. Erdmann, R. G. Ulbrich, M. Grochol, F. Grosse, R. Zimmermann, S. Malzer, and G. H. Döhler, *ibid.* **75**, 115317 (2007).
[6] I. V. Ponomarev, L. I. Deych, and A. A. Lisyansky, Phys. Rev. B **71**, 155303 (2005).
[7] J. B. B. de Oliveira, E. A. Meneses, and E. C. F. da Silva, Phys. Rev. B **60**, 1519 (1999).
[8] I. Aksenov, J. Kusano, Y. Aoyagi, T. Sugano, T. Yasuda, and Y. Segawa, Phys. Rev. B **51**, 4278 (1995).
[9] A. Polimeni, R. K. Hayden, L. Eaves, M. Henini, P. C. Main, K. Uchida, N. Miura, J. Main, and G. Wunner, Physica E (Amsterdam) **13**, 49 (2002).
[10] H. Sasaki, Y. Arakawa, M. Nishioka, J. Yoshino, H. Okamoto, and N. Miura, Appl. Phys. Lett. **46**, 83 (1985).
[11] K. Vahala, Y. Arakawa, and A. Yariv, Appl. Phys. Lett. **50**, 365 (1987).